

### **Listing of The Claims**

Please cancel claims 1-50.

51. (Currently Amended) A method for MRI imaging comprising:  
administering to a patient an MRI contrast agent, comprising a perfluoroalkyl-containing metal complex that has a critical micelle formation concentration  $< 10^{-3}$  mol/l, a hydrodynamic micelle diameter ( $2 R_h$ )  $> 1$  nm and a proton relaxivity in plasma ( $R^1$ )  $> 10$  l/mmol's ,

allowing the uptake of contrast agent in tissue ,

conducting MRI imaging,

and visualizing whereby plaque in which contrast agent is uptaken, infarcted tissue, or necrotic tissue are visualized

or

independently simultaneously visualizing necroses and tumors in which contrast agent is uptaken are independently visualized.

52. (Canceled)

53. (Previously Presented) A method according to claim 51, wherein necroses or tumors are independently visualized.

54. (Previously Presented) A method according to claim 51, wherein the metal complex has a micelle formation concentration of  $< 10^{-4}$  mol/l.

55. **(Previously Presented)** A method according to claim 51, wherein the metal complex has a hydrodynamic micelle diameter of > 3 nm.

56. **(Previously Presented)** A method according to claim 51, wherein the metal complex has a proton relaxivity in plasma of > 13 l/mmol's.

57. **(Currently amended)** A method according to claim 51, wherein the perfluoroalkyl-containing metal complex is a compound of formula I

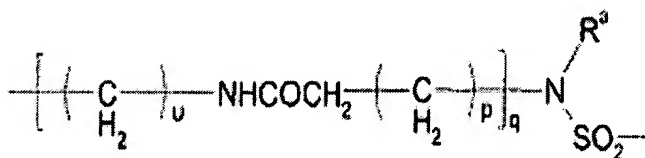


in which

$R^F$  is a perfluorinated, straight-chain or branched carbon chain with formula - $C_nF_{2n}E$ , in which

$E$  is a terminal fluorine, chlorine, bromine, iodine or hydrogen atom and  
 $n$  is a number from 4-30,

$L$  is a direct bond, a methylene group, an -NHCO- group, a group



whereby  $p$  is a number from 0 to 10, and  $q$  and  $n$ , independently of one another, are 0 or 1, and  $R^a$  is a hydrogen atom, a methyl group, a -CH<sub>2</sub>-OH group, a -CH<sub>2</sub>-CO<sub>2</sub>H group or a C<sub>2</sub>-C<sub>15</sub> alkyl, which optionally is interrupted by 1 to 3 oxygen atoms, 1 to 2 CO groups or an optionally substituted aryl group and/or is substituted with 1 to 4 hydroxyl groups, 1 to 2

C<sub>1</sub>-C<sub>4</sub> alkoxy groups, 1 to 2 carboxy groups, or a group -SO<sub>3</sub>H,

or

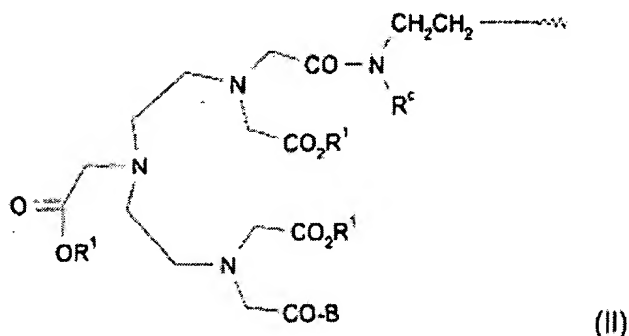
L is a straight-chain, branched, saturated or unsaturated C<sub>2</sub>-C<sub>30</sub> carbon chain, which optionally contains 1 to 10 oxygen atoms, 1 to 3 -NR<sup>a</sup> groups, 1 to 2 sulfur atoms, a piperazine group, a -CONR<sup>a</sup> group, an -NR<sup>a</sup>CO group, an -SO<sub>2</sub> group, an -NR<sup>a</sup>-CO<sub>2</sub> group, 1 to 2 CO groups, a group -CO-N-T-N(R<sup>a</sup>)-SO<sub>2</sub>-R<sup>F</sup>, or 1 to 2 optionally substituted aryls and/or is interrupted by these groups and/or is optionally substituted with 1 to 3 -OR<sup>a</sup> groups, 1 to 2 oxo groups, 1 to 2 -NH-COR<sup>a</sup> groups, 1 to 2 -CONHR<sup>a</sup> groups, 1 to 2 -(CH<sub>2</sub>)<sub>p</sub>-CO<sub>2</sub>H groups, 1 to 2 groups -(CH<sub>2</sub>)<sub>p</sub>-(O)<sub>q</sub>-CH<sub>2</sub>CH<sub>2</sub>-R<sup>F</sup>,

whereby

R<sup>a</sup>, R<sup>F</sup> and p and q have the above-indicated meanings, and

T is a C<sub>2</sub>-C<sub>10</sub> chain, which optionally is interrupted by 1 to 2 oxygen atoms or 1 to 2 -NHCO groups,

K is a complexing agent or metal complex of formula II



in which R<sup>c</sup>, R<sup>1</sup> and B are independent of one another,

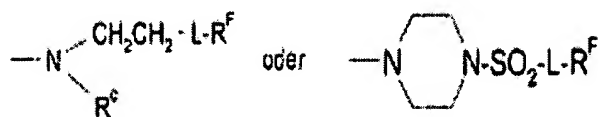
and

R<sup>c</sup> is R<sup>a</sup> or is -(CH<sub>2</sub>)<sub>m</sub>-L-R<sup>F</sup>, whereby m is 0, 1 or 2, and L and R<sup>F</sup> have the above-mentioned meaning,

R<sup>1</sup>, independently of one another, is a hydrogen atom or a metal ion equivalent

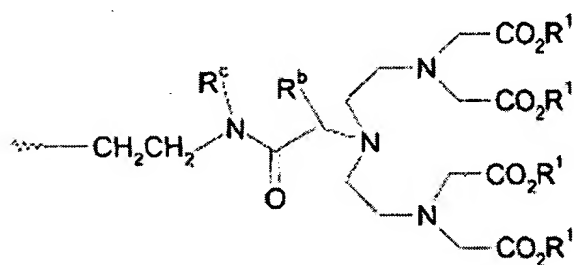
of atomic numbers 22-29, 42-46 or 58-70,

B is  $-OR^1$ ,



whereby  $R^1$ , L,  $R^F$  and  $R^c$  have the above-mentioned meanings, or

K is a complexing agent or complex of formula III

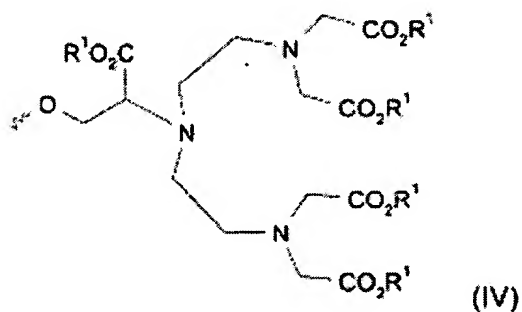


(III)

in which  $R^c$  and  $R^1$  have the above-mentioned meanings and  $R^b$  has the meaning of  $R^a$

or

K is a complexing agent or complex of formula IV

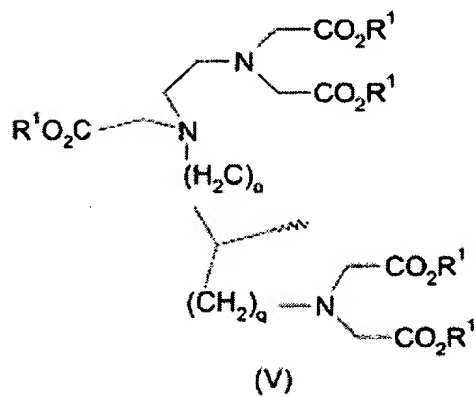


(IV)

in which  $R^1$  has the above-mentioned meaning

or

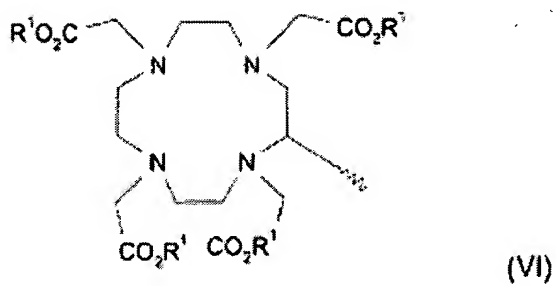
K is a complexing agent or complex of formula V



in which R<sup>1</sup> has the above-mentioned meaning, and o and q stand for numbers 0 or 1, and yields the sum o + q = 1,

or

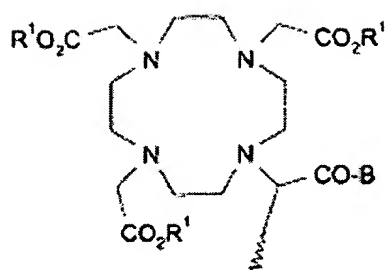
K is a complexing agent or complex of formula VI



in which R<sup>1</sup> has the above-mentioned meaning

or

K is a complexing agent or complex of formula VII

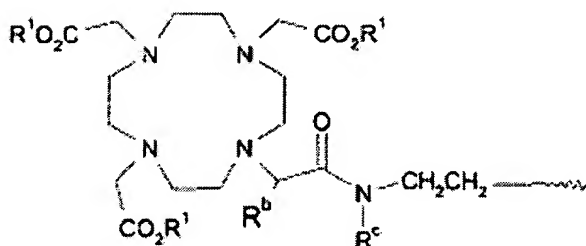


(VII)

in which R<sup>1</sup> and B have the above-mentioned meanings

or

K is a complexing agent or complex of formula VIII

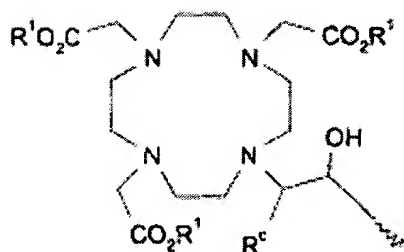


(VIII)

in which R<sup>c</sup> and R<sup>1</sup> have the above-mentioned meanings, and R<sup>b</sup> is R<sup>a</sup>

or

K is a complexing agent or complex of formula IX

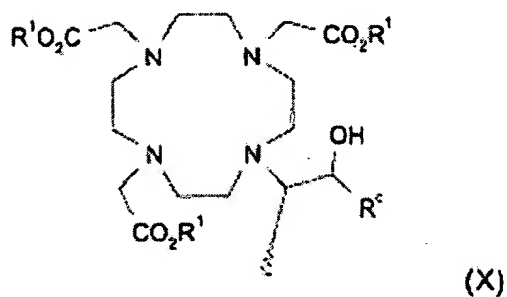


(IX)

in which R<sup>c</sup> and R<sup>1</sup> have the above-mentioned meanings,

or

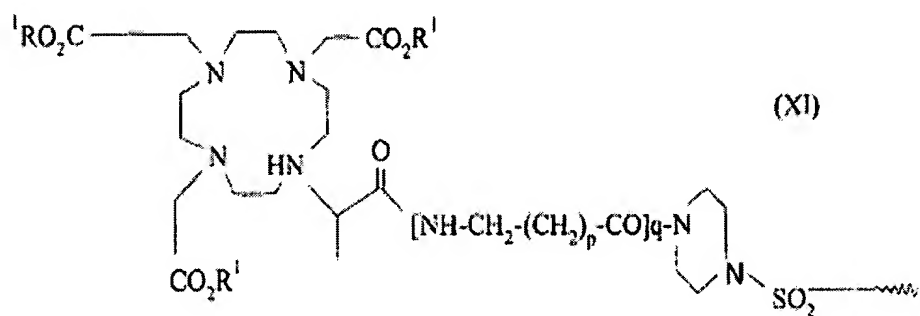
K is a complexing agent or complex of formula X



in which  $R^c$  and  $R^1$  have the above-mentioned meanings,

or

K is a complexing agent or complex of formula XI

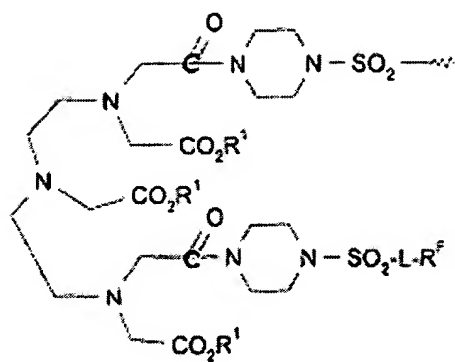


in which  $R^1$ ,  $p$  and  $q$  have the above-mentioned meanings,

and  $R^b$  has the meaning of  $R^a$ ,

or

K is a complexing agent or complex of formula XII

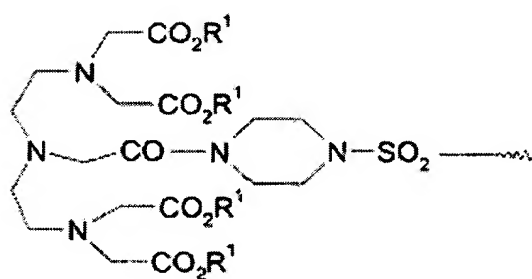


(XII)

in which L, R<sup>F</sup> and Z<sup>1</sup> have the above-mentioned meanings,

or

K is a complexing agent or complex of formula XIII



(XIII)

in which R<sup>1</sup> has the above-mentioned meaning, or

K is a salt of one of the complexing agents or complexes of formula II to XIII with an organic and/or inorganic base or amino acid or amino acid amide.

58. **(Previously Presented)** A method according to claim 57, wherein in the compound of formula I,

L is

$\alpha$ -CH<sub>2</sub>- $\beta$

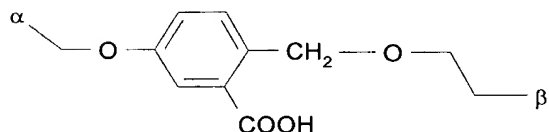
$\alpha$ -CH<sub>2</sub>CH<sub>2</sub>- $\beta$

$\alpha$ -(CH<sub>2</sub>)<sub>s</sub>- $\beta$  s = 3 - 15



$\alpha\text{-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-(O-CH}_2\text{-CH}_2\text{)}_t\text{-}\beta \quad t = 2 - 6$   
 $\alpha\text{-CH}_2\text{-NH-CO-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(CH}_2\text{COOH)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(C}_{10}\text{H}_{21}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(C}_6\text{H}_{13}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-(CH}_2\text{)}_{10}\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-CH}_2\text{-OH)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NHCO-(CH}_2\text{)}_{10}\text{-S-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{NHCOCH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{NHCO(CH}_2\text{)}_{10}\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-C}_6\text{H}_4\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-O-CH}_2\text{-C(CH}_2\text{-OCH}_2\text{CH}_2\text{-C}_6\text{F}_{13}\text{)}_2\text{-CH}_2\text{-OCH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NHCOCH}_2\text{CH}_2\text{CON-CH}_2\text{CH}_2\text{NHCOCH}_2\text{N(C}_2\text{H}_5\text{)SO}_2\text{C}_8\text{F}_{17}$

$\text{CH}_2\text{-CH}_2\text{NHCOCH}_2\text{N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-O-CH}_2\text{-CH(OC}_{10}\text{H}_{21}\text{)-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-(CH}_2\text{NHCO)}_4\text{-CH}_2\text{O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-(CH}_2\text{NHCO)}_3\text{-CH}_2\text{O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-OCH}_2\text{C(CH}_2\text{OH)}_2\text{-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$



$\alpha\text{-CH}_2\text{NHCOCH}_2\text{N(C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NHCO-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-NHCO-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(CH}_2\text{COOH)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_{10}\text{H}_{21}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_6\text{H}_{13}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-(CH}_2\text{)}_{10}\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$

$\alpha$ -NH-CO-CH<sub>2</sub>-N(-CH<sub>2</sub>-CH<sub>2</sub>-OH)SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-O-C<sub>6</sub>H<sub>4</sub>-O-CH<sub>2</sub>-CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-C<sub>6</sub>H<sub>4</sub>-O-CH<sub>2</sub>-CH<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>2</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>10</sub>H<sub>21</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>6</sub>H<sub>13</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>2</sub>H<sub>4</sub>OH)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(CH<sub>2</sub>COOH)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(CH<sub>2</sub>C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N-[CH(CH<sub>2</sub>OH)<sub>2</sub>]-SO<sub>2</sub>- $\beta$  or  
 $\alpha$ -N-[CH(CH<sub>2</sub>OH)CH(CH<sub>2</sub>OH)]-SO<sub>2</sub>- $\beta$ ,

in which  $\alpha$  is the binding site to the complexing agent or metal complex K, and  $\beta$  is the binding site to the fluorine radical.

59. **(Previously Presented)** A method according to claim 57, wherein the compound of formula I, is a compound in which n in formula -C<sub>n</sub>F<sub>2n</sub>E is a number from 4-15 and/or E is a fluorine atom.

60. **(Previously Presented)** A method according to claims 57, wherein the compound of formula I is:

Gadolinium complex of 10-[1-methyl-2-oxo-3-aza-5-oxo-{4-perfluorooctylsulfonyl-piperazin-1-yl}-pentyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,  
 Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-oxa-10,10,11,11,12,12,13,13,14,14,15,15,16,16,17,17-heptafluoroheptadecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,  
 Gadolinium complex of 10-[2-hydroxy-4-aza-5,9-dioxo-9-{4-perfluorooctyl}-piperazin-1-yl]-nonyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-aza-7-(perfluorooctyl-sulfonyl)-nonyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,  
 Gadolinium complex of 10-[2-hydroxy-4-oxa-1H,1H,2H,3H,3H,5H,5H,6H,6H-perfluorotetradecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,  
 Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-oxa-10,10,11,11,12,12,13,13,14,14,15,15,-16,16,17,17,18,18,19,19-henicosafuoro-nonadecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,  
 Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-11-aza-11-(perfluorooctylsulfonyl)-tridecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane, or  
 Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-aza-7-(perfluorooctylsulfonyl)-8-phenyl-octyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraaza-cyclododecane.

61. **(Previously Presented)** A method according to claim 51, wherein the perfluoroalkyl-containing metal complex, is a compound of formula Ia



in which

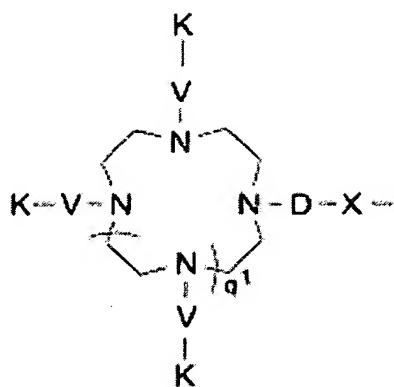
A is a group that contains 2 to 6 metal complexes, which are bonded directly or via a linker to a nitrogen atom of an annular skeleton chain,  
 and

$R^F$  is a perfluorinated, straight-chain or branched carbon chain with formula  $-C_nF_{2n}E$ , in which

E is a terminal fluorine, chlorine, bromine, iodine or hydrogen atom,

and n is a number from 4-30,

whereby A has the following structure:



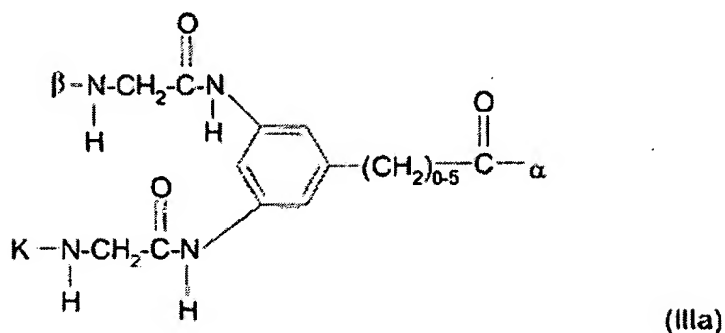
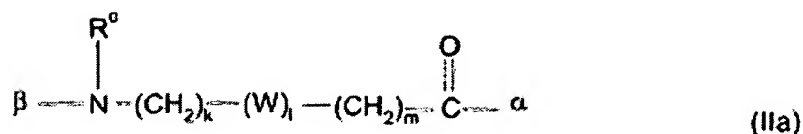
whereby

$q^1$  is 0, 1, 2 or 3,

K is a complexing agent or metal complex or a salts thereof with an organic and/or inorganic base or amino acid or amino acid amide,

X as the point of attachment to  $R^F$ , is a direct bond, a phenylene group or a  $C_1$ - $C_{10}$  alkylene chain, which optionally contains 1-15 oxygen atoms, 1-5 sulfur atoms, 1-10 carbonyl groups, 10-10 ( $NR^d$ ) groups, 1-2  $NR^dSO_2$  groups, 1-10  $CONR^d$  groups, 1 piperidine group, 1-3  $SO_2$  groups and/or 1-2 phenylene groups or optionally is substituted by 1-3 radicals  $R^F$ , in which  $R^d$  is a hydrogen atom, a phenyl group, benzyl group or a  $C_1$ - $C_{15}$  alkyl group, which optionally contains 1-2  $NHCO$ , 1-2  $CO$  groups, 1-5 oxygen atoms and optionally is substituted by 1-5 hydroxy, 1-5 methoxy, 1-3 carboxy, or 1-3  $R^F$  radicals,

V is a direct bond or a chain of formula IIa or IIIa:



in which

- $\text{R}^e$  is a hydrogen atom, a phenyl group, a benzyl group or a  $\text{C}_1\text{-C}_7$  alkyl group, which optionally is substituted with a carboxy group, a methoxy group or a hydroxy group,
- $\text{W}$  is a direct bond, a polyglycol ether group with up to 5 glycol units, or a group of formula IVa



in which  $\text{R}^h$  is a  $\text{C}_1\text{-C}_7$  carboxylic acid, a phenyl group, a benzyl group or a  $(\text{CH}_2)_{1-5}\text{-NH-K}$  group,

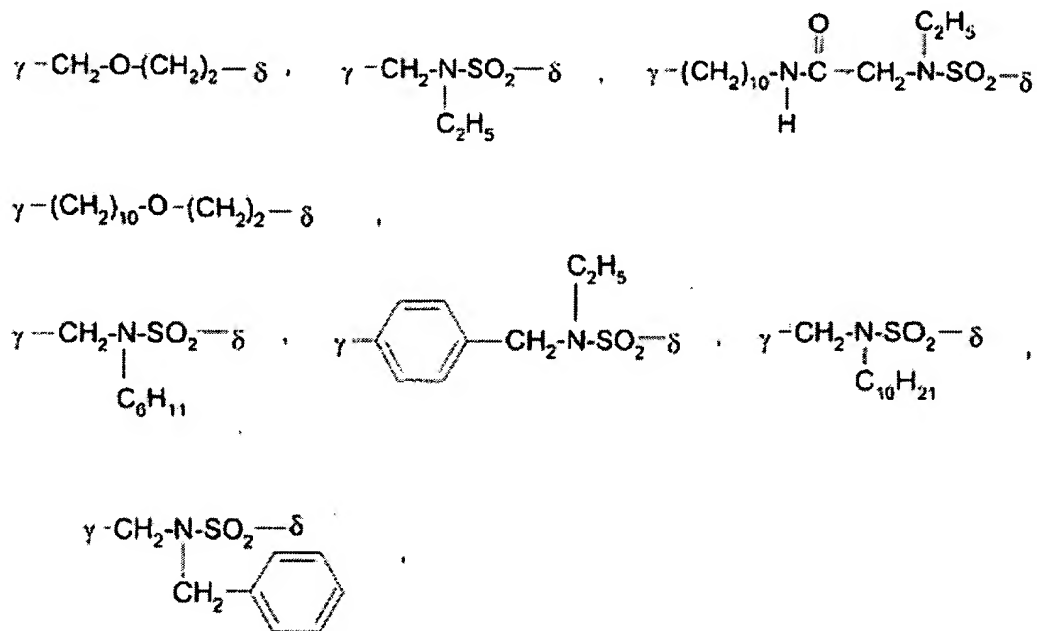
- $\alpha$  is the binding to the nitrogen atom of the skeleton chain,  $\beta$  is the binding to complexing agent or metal complex  $\text{K}$ ,
- and in which variables  $k$  and  $m$  stand for natural numbers between 0 and 10, and  $l$  is 0 or 1

and whereby

$\text{D}$  is a  $\text{CO}$  or  $\text{SO}_2$  group.

62. **(Previously Presented)** A method according to claim 61, wherein the compound of formula Ia is a compound in which  $q^1$  is the number 1.

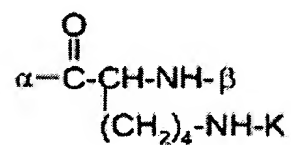
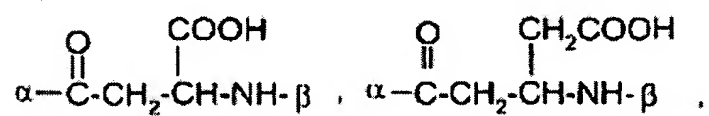
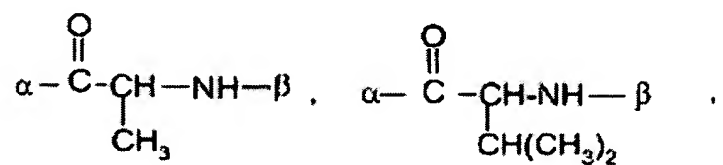
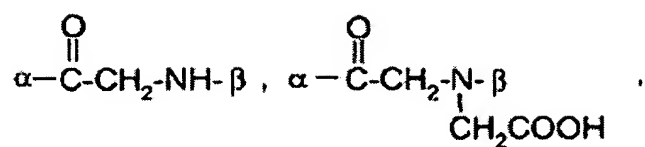
63. **(Previously Presented)** A method according to claim 61, wherein the compound of formula Ia is a compound in which X is an alkylene chain, which contains 1-10 -CH<sub>2</sub>CH<sub>2</sub>O- groups or 1-5 -COCH<sub>2</sub>NH- groups, a direct bond or one of the following structures



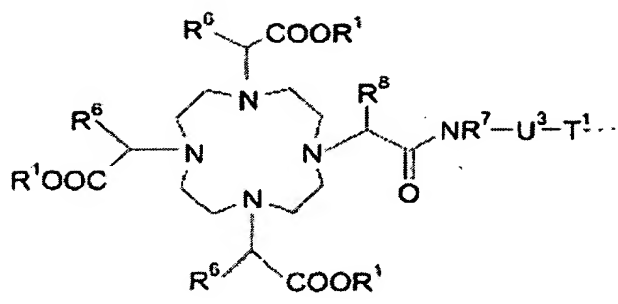
whereby

$\gamma$  binds to D, and  $\delta$  binds to R<sup>F</sup>.

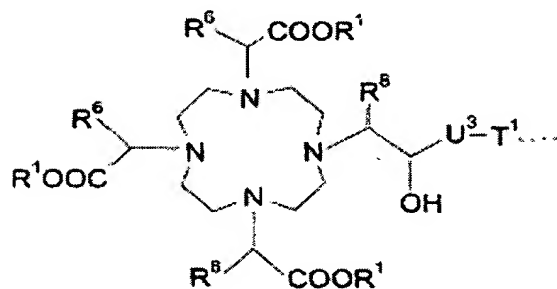
64. **(Previously Presented)** A method according to claim 61, wherein the compound of formula Ia, is a compound in which V is a group with one of the following structures



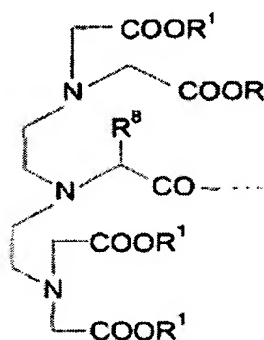
65. **(Previously Presented)** A method according to claim 61, wherein the compound of formula Ia, is a compound in which K is a complexing agent or complex of formula Va, VIa, VIIa or VIIIa,



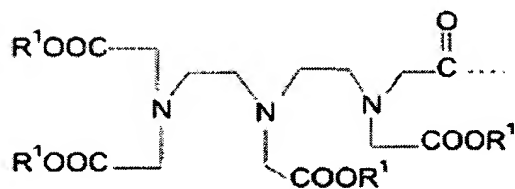
(Va)



(Via)



(VIIa)



(VIIIa)

whereby

$\text{R}^1$ , independently of one another, are a hydrogen atom or a metal ion equivalent of the elements of atomic numbers 23-29, 42-46 or 58-70,

$\text{R}^8$  is a hydrogen atom or a straight-chain, branched, saturated or unsaturated  $\text{C}_1\text{-C}_{30}$  alkyl chain, which optionally is substituted by 1-5 hydroxy, 1-3



carboxy or 1 phenyl group(s) and/or optionally is interrupted by 1-10 oxygen atoms, 1 phenylene group or 1 phenylenoxy group,

$R^6$  are independently a hydrogen atom, a straight-chain or branched  $C_1$ - $C_7$  alkyl radical, a phenyl radical or benzyl radical,

$R^7$  is a hydrogen atom, a methyl group or ethyl group, which optionally is substituted by a hydroxy group or carboxy group,

$U^3$  is a straight-chain, branched, saturated or unsaturated  $C_1$ - $C_{20}$  alkylene group optionally containing 1-5 imino groups, 1-3 phenylene groups, 1-3 phenylenoxy groups, 1-3 phenylenimino groups, 1-5 amide groups, 1-2 hydrazide groups, 1-5 carbonyl groups, 1-5 ethylenoxy groups, 1 urea group, 1 thiourea group, 1-2 carboxyalkylimino groups, 1-2 ester groups, 1-1-0 oxygen atoms, 1-5 sulfur atoms and/or 1-5 nitrogen atoms, and/or optionally substituted by 1-5 hydroxy groups, 1-2 mercapto groups, 1-5 oxo groups, 1-5 thioxo groups, 1-3 carboxy groups, 1-5 carboxyalkyl groups, 1-5 ester groups and/or 1-3 amino groups, whereby the optionally contained phenylene groups can be substituted by 1-2 carboxy groups, 1-2 sulfone groups or 1-2 hydroxy groups

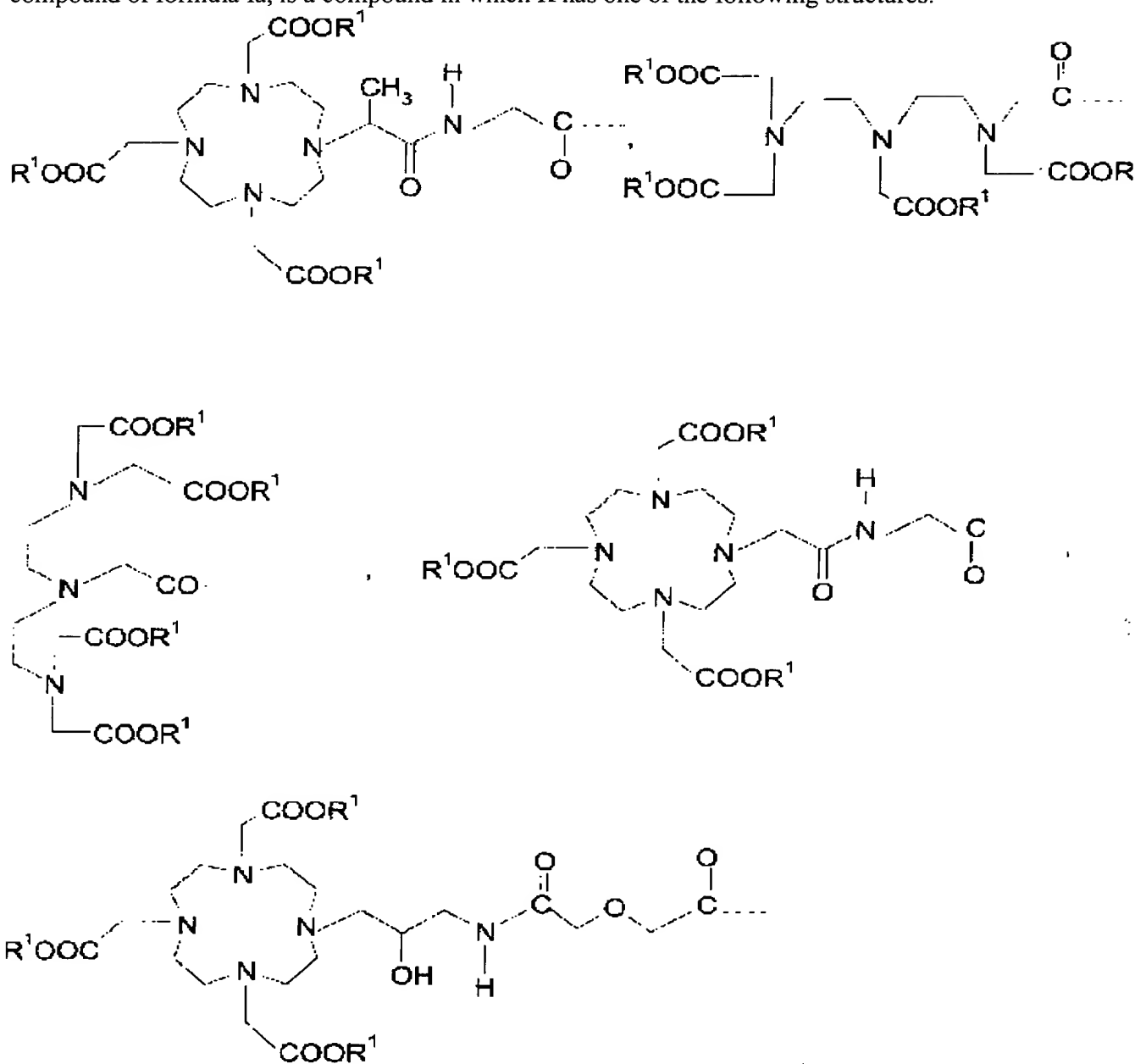
$T^1$  is a  $-CO-\beta$ ,  $-NHCO-\beta$  or  $-NHCS-\beta$  group, whereby  $\beta$  is the binding site to V.

66. **(Previously Presented)** A method according to claim 65, wherein the  $C_1$ - $C_{20}$  alkylene chain that is  $U^3$  contains the group  $-CH_2NHCO-$ ,  $-NHCOCH_2O-$ ,  $-NHCOCH_2OC_6H_4-$ ,  $-N(CH_2CO_2H)-$ ,  $-CH_2OCH_2-$ ,  $-NHCOCH_2C_6H_4-$ ,  $-NHCSNHC_6H_4-$ ,  $-CH_2OC_6H_4-$ , or  $-CH_2CH_2O-$  and/or is substituted by the group  $-COOH$  and/or  $-CH_2COOH$ .

67. **(Previously Presented)** A method according to claim 65, wherein  $U^3$  is a  $-CH_2-$ ,  $-CH_2CH_2-$ ,  $-CH_2CH_2CH_2-$ ,  $-C_6H_4-$ ,  $-C_6H_{10}-$ ,  $-CH_2C_6H_4-$ ,  $-CH_2NHCOCH_2CH(CH_2CO_2H)-C_6H_4-$ ,  $-CH_2NHCOCH_2OCH_2-$ , or  $-CH_2NHCOCH_2C_6H_4-$

group.

68. **(Previously Presented)** A method according to claim 61, wherein the compound of formula Ia, is a compound in which K has one of the following structures:



69. **(Previously Presented)** A method according to claim 61 , wherein the compound of formula Ia, is a compound in which the perfluoroalkyl chain is  $R^F$  is  $-C_6F_{13}$ ,  $-C_8F_{17}$ ,  $-C_{10}F_{21}$  or  $-C_{12}F_{25}$  .

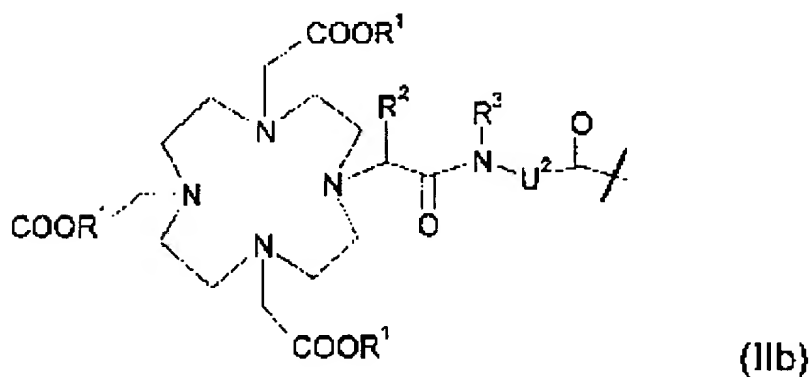
70. **(Previously Presented)** A method according to claim 61 , wherein the compound of formula Ia is a gadolinium complex of 1,4,7-tris{1,4,7-tris(N-(carboxylatomethyl)-10-[N-1-methyl-3,6-diaza-2,5,8-trioxooctane-1,8-diyl])-1,4,7,10-tetraazacyclododecane, Gd complex}-10-[N-2H,2H,4H,4H,5H,5H-3-oxa-perfluorotridecanoyl]-1,4,7,10-tetraazacyclododecane .

71. **(Previously Presented)** A method according to claim 51 , wherein the perfluoroalkyl-containing metal complex, is a compound of formula Ib



in which

K is a complexing agent or a metal complex of formula IIb



whereby

$R^1$  is a hydrogen atom or a metal ion equivalent of atomic numbers 23-

29, 42-46 or 58-70,

$R^2$  and  $R^3$  are independently a hydrogen atom, a  $C_1$ - $C_7$  alkyl group, a benzyl group, a phenyl group,  $-CH_2OH$  or  $-CH_2-OCH_3$ ,

$U^2$  is radical  $L^1$ , whereby  $L^1$  and  $U^2$ , independently of one another, can be the same or different,

$A^1$  is a hydrogen atom, a straight-chain or branched  $C_1$ - $C_{30}$  alkyl group, which optionally is interrupted by 1-15 oxygen atoms, and/or optionally is substituted with 1-10 hydroxy groups, 1-2  $COOH$  groups, a phenyl group, a benzyl group and/or 1-5  $-OR^9$  groups, with  $R^9$  having the meaning of a hydrogen atom or a  $C_1$ - $C_7$  alkyl radical, or  $-L^1-R^F$ ,

$L^1$  is a straight-chain or branched  $C_1$ - $C_{30}$  alkylene group, which optionally is interrupted by 1-10 oxygen atoms, 1-5  $-NH-CO$  groups, 1-5  $-CO-NH$  groups, by a phenylene group optionally substituted by a  $COOH$ - group, 1-3 sulfur atoms, 1-2  $-N(B^1)-SO_2$  groups and/or 1-2  $-SO_2-N(B^1)$ -groups with  $B^1$  in the meaning of  $A^1$ , an  $NHCO$  group, a  $CONH$  group, an  $N(B^1)-SO_2$  group or an  $-SO_2-N(B^1)$  group and/or optionally is substituted with radical  $R^F$ , and

$R^F$  is a straight-chain or branched perfluorinated alkyl radical of formula  $C_nF_{2n}E$ , whereby  $n$  is number 4-30, and

$E$  is a terminal fluorine atom, chlorine atom, bromine atom, iodine atom or a hydrogen atom,

and optionally present acid groups optionally can be present as salts of organic and/or inorganic bases or amino acids or amino acid amides.

**72. (Previously Presented)** A method according to claim 71, wherein the compound of formula Ib, is a compound in which  $R^2$ ,  $R^3$  and  $R^9$ , independently of one another, mean hydrogen or a  $C_1$ - $C_4$  alkyl group.

**73. (Previously Presented)** A method according to claim 71, wherein the

compound of formula Ib, is a compound in which A<sup>1</sup> is hydrogen, a C<sub>1</sub>-C<sub>5</sub> alkyl radical, or the radicals

C<sub>2</sub>H<sub>4</sub>-O-CH<sub>3</sub>, C<sub>3</sub>H<sub>6</sub>-O-CH<sub>3</sub>,  
 C<sub>2</sub>H<sub>4</sub>-O-(C<sub>2</sub>H<sub>4</sub>-O)<sub>t</sub>-C<sub>2</sub>H<sub>4</sub>-OH,  
 C<sub>2</sub>H<sub>4</sub>-O-(C<sub>2</sub>H<sub>4</sub>-O)<sub>t</sub>-C<sub>2</sub>H<sub>4</sub>-OCH<sub>3</sub>, C<sub>2</sub>H<sub>4</sub>OH,  
 C<sub>3</sub>H<sub>6</sub>OH, C<sub>4</sub>H<sub>8</sub>OH, C<sub>5</sub>H<sub>10</sub>OH, C<sub>6</sub>H<sub>12</sub>OH, C<sub>7</sub>H<sub>14</sub>OH,  
 CH(OH)CH<sub>2</sub>OH,  
 CH(OH)CH(OH)CH<sub>2</sub>OH, CH<sub>2</sub>[CH(OH)]<sub>u</sub><sup>1</sup>CH<sub>2</sub>OH,  
 CH[CH<sub>2</sub>(OH)]CH(OH)CH<sub>2</sub>OH,  
 C<sub>2</sub>H<sub>4</sub>CH(OH)CH<sub>2</sub>OH,  
 (CH<sub>2</sub>)<sub>s</sub>COOH,  
 C<sub>2</sub>H<sub>4</sub>-O-(C<sub>2</sub>H<sub>4</sub>-O)<sub>t</sub>-CH<sub>2</sub>COOH , or  
 C<sub>2</sub>H<sub>4</sub>-O-(C<sub>2</sub>H<sub>4</sub>-O)<sub>t</sub>-C<sub>2</sub>H<sub>4</sub>-C<sub>n</sub>F<sub>2n</sub>E

whereby

s is integers 1 to 15,

t is integers 0 to 13,

u<sup>1</sup> is integers 1 to 10,

n is integers 4 to 20, and

E is hydrogen, fluorine, chlorine, bromine or iodine atoms, and optionally, their

branched isomers.

74. **(Previously Presented)** A method according to claim 71, wherein the compound of formula Ib, is a compound in which A<sup>1</sup> is hydrogen, C<sub>1</sub>-C<sub>10</sub> alkyl,  
 C<sub>2</sub>H<sub>4</sub>-O-CH<sub>3</sub>, C<sub>3</sub>H<sub>6</sub>-O-CH<sub>3</sub>,  
 C<sub>2</sub>H<sub>4</sub>-O-(C<sub>2</sub>H<sub>4</sub>-O)<sub>x</sub>-C<sub>2</sub>H<sub>4</sub>-OH, C<sub>2</sub>H<sub>4</sub>-O-(C<sub>2</sub>H<sub>4</sub>-O)<sub>x</sub>-C<sub>2</sub>H<sub>4</sub>-OCH<sub>3</sub>,  
 C<sub>2</sub>H<sub>4</sub>OH, C<sub>3</sub>H<sub>6</sub>OH,

$\text{CH}_2[\text{CH}(\text{OH})]_y\text{CH}_2\text{OH}$ ,  
 $\text{CH}[\text{CH}_2(\text{OH})]\text{CH}(\text{OH})\text{CH}_2\text{OH}$ ,  
 $(\text{CH}_2)_w\text{COOH}$ ,  
 $\text{C}_2\text{H}_4\text{-O-(C}_2\text{H}_4\text{-O)}_x\text{-CH}_2\text{COOH}$  or  
 $\text{C}_2\text{H}_4\text{-O-(C}_2\text{H}_4\text{-O)}_x\text{-C}_2\text{H}_4\text{-C}_n\text{F}_{2n}\text{E}$ ,

whereby

x is integers 0 to 5,

y is integers 1 to 6,

w is integers 1 to 10,

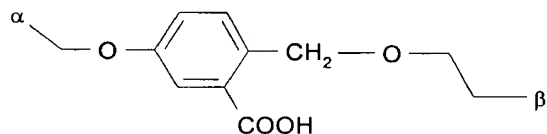
n is integers 4 to 15, and

E is a fluorine atom, and, optionally, their branched isomers.

75. **(Previously Presented)** A method according to claim 71, wherein the compound of formula Ib, is a compound in which  $\text{L}^1$  is

$\alpha\text{-(CH}_2)_8\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-CH}_2\text{-(O-CH}_2\text{-CH}_2\text{)}_y\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-(O-CH}_2\text{-CH}_2\text{)}_y\text{-}\beta$ ,  
 $\alpha\text{-CH}_2\text{-NH-CO-}\beta$   
 $\alpha\text{-CH}_2\text{-CH}_2\text{-NH-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(CH}_2\text{COOH)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(C}_{10}\text{H}_{21}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(C}_6\text{H}_{13}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-(CH}_2\text{)}_{10}\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-CH}_2\text{-OH)SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NHCO-(CH}_2\text{)}_{10}\text{-S-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{NHCOCH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-CH}_2\text{NHCOCH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-(CH}_2\text{-CH}_2\text{-O)}_t\text{-(CH}_2\text{)}_3\text{NHCO-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{NHCO(CH}_2\text{)}_{10}\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{CH}_2\text{NHCO(CH}_2\text{)}_{10}\text{-O-CH}_2\text{CH}_2\text{-}\beta$

$\alpha\text{-CH}_2\text{-C}_6\text{H}_4\text{-O-CH}_2\text{CH}_2\text{-}\beta$  whereby the phenylene group 1,4 or 1,3 is linked  
 $\alpha\text{-CH}_2\text{-O-CH}_2\text{-C(CH}_2\text{-OCH}_2\text{CH}_2\text{-C}_6\text{F}_{13})_2\text{-CH}_2\text{-OCH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-NHCOCH}_2\text{CH}_2\text{CON-CH}_2\text{CH}_2\text{NHCOCH}_2\text{N(C}_2\text{H}_5\text{)SO}_2\text{C}_8\text{F}_{17}\beta$   
 $\alpha\text{-CH}_2\text{-CH}_2\text{NHCOCH}_2\text{N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-O-CH}_2\text{-CH(OC}_{10}\text{H}_{21}\text{)-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-(CH}_2\text{NHCO)}_4\text{-CH}_2\text{O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-(CH}_2\text{NHCO)}_3\text{-CH}_2\text{O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-OCH}_2\text{C(CH}_2\text{OH)}_2\text{-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$



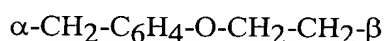
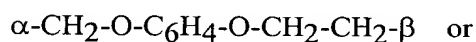
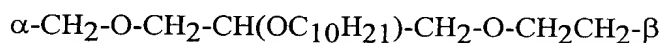
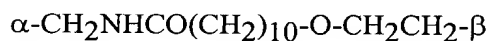
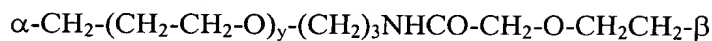
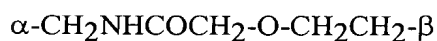
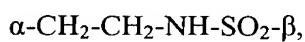
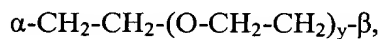
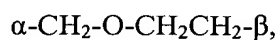
$\alpha\text{-CH}_2\text{NHCOCH}_2\text{N(C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NHCO-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-NHCO-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(CH}_2\text{COOH)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_{10}\text{H}_{21}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_6\text{H}_{13}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-(CH}_2\text{)}_{10}\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-CH}_2\text{-OH)SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-O-C}_6\text{H}_4\text{-O-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-C}_6\text{H}_4\text{-O-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_{10}\text{H}_{21}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_6\text{H}_{13}\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_2\text{H}_4\text{OH)-SO}_2\text{-}\beta$   
 $\alpha\text{-N(CH}_2\text{COOH)-SO}_2\text{-}\beta$   
 $\alpha\text{-N(CH}_2\text{C}_6\text{H}_5\text{)-SO}_2\text{-}\beta$   
 $\alpha\text{-N-[CH(CH}_2\text{OH)}_2\text{]-SO}_2\text{-}\beta$  or  
 $\alpha\text{-N-[CH(CH}_2\text{OH)CH(OH)(CH}_2\text{OH)]-SO}_2\text{-}\beta$

whereby

s is integers 1 to 15 and

y is integers 1 to 6.

76. **(Previously Presented)** A method according to claim 71, wherein the compound of formula Ib, is a compound in which L<sup>1</sup> is



whereby

y is an interger from 1 to 6.

77. **(Previously Presented)** A method according to claim 71, wherein the compound of formula Ib, is a compound in which R<sup>F</sup> is a straight-chain or branched perfluorinated alkyl radical of formula C<sub>n</sub>F<sub>2n</sub>E, whereby n is a number from 4 to 15 and E stands for a terminal fluorine atom.



78. **(Previously Presented)** A method according to claim 71 , wherein the compound of formula Ib is a :

1,4,7-Tris(carboxylatomethyl)-10-(3-aza-4-oxo-hexan-5-yl)-acid-(2,3-dihydroxypropyl)-N-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide]-1,4,7,10-tetraazacyclododecane, gadolinium complex,

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)acid-N-(3,6,9,12,15-pentaoxa)-hexadecyl)-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide}-1,4,7,10-tetraazacyclododecane, gadolinium complex,

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-N-5-hydroxy-3-oxa-pentyl)-N-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide}-1,4,7,10-tetraazacyclododecane, gadolinium complex,

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-[N-3,6,9,15-tetraoxa-12-aza-15-oxo-C<sub>17</sub>-C<sub>26</sub>-hepta-decafluor)hexacosyl]-amide}-1,4,7,10-tetraazacyclododecane, gadolinium complex, or

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-N-(2-methoxyethyl)-N-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide}-1,4,7,10-tetraazacyclododecane, gadolinium complex.

79. **(Previously Presented)** A method according to claim 57, wherein the perfluoroalkyl-containing metal complex is in a galenical formulation that contains a paramagnetic, perfluoroalkyl-containing metal complex of formula I, and a diamagnetic perfluoroalkyl-containing substance, optionally dissolved in an aqueous solvent, wherein the diamagnetic perfluoroalkyl-containing substance is a compound of formula XX



in which  $R^F$  is a straight-chain or branched perfluoroalkyl radical with 4 to 30 carbon atoms,  $L^2$  is a linker and  $B^2$  is a hydrophilic group.

80. **(Previously Presented)** A method according to claim 79, wherein linker  $L^2$  is a direct bond, an  $-SO_2$  group, or a straight-chain or branched carbon chain with 1 to 20 carbon atoms, which can be substituted with one or more  $-OH$ ,  $-COO$ ,  $-SO_3$  groups and/or optionally contains one or more  $-O-$ ,  $-S-$ ,  $-CO-$ ,  $-CONH-$ ,  $-NHCO-$ ,  $-CONR^9-$ ,  $-NR^9CO-$ ,  $-SO_2-$ ,  $-PO_4-$ ,  $-NH-$  or  $-NR^9$  groups, an aryl ring or a piperazine, whereby  $R^9$  is a  $C_1$  to  $C_{20}$  alkyl radical, which in turn can contain one or more O atoms, and/or can be substituted with  $-COO-$  or  $SO_3$  groups.

81. **(Previously Presented)** A method according to claim 79, wherein hydrophilic group  $B^2$  is a mono- or disaccharide, with one or more adjacent  $-COO^-$  or  $-SO_3$  groups, a dicarboxylic acid, an isophthalic acid, a picolinic acid, a benzenesulfonic acid, a tetrahydropyrandicarboxylic acid, a 2,6-pyridinedicarboxylic acid, a quaternary ammonium ion, an aminopolycarboxylic acid, an aminodipolyethylene glycolsulfonic acid, an aminopolyethylene glycol group, an  $SO_2-(CH_2)_2-OH$  group, a polyhydroxyalkyl chain with at least two hydroxyl groups or one or more polyethylene glycol chains with at least two glycol units, whereby the polyethylene glycol chains are terminated by an  $-OH$  or  $-OCH_3$  group.

82. **(Previously presented)** A method according to claim 55, wherein the metal complex has a hydrodynamic micelle diameter of  $> 4$  nm.

83. **(Previously presented)** A method according to claim 56, wherein the metal complex has a proton relaxivity in plasma of  $> 15$  l/mmol's.

84. **(Previously presented)** A method according to claim 61, wherein the perfluoroalkyl-containing metal complex is in a galenical formulation that contains a

paramagnetic, perfluoroalkyl-containing metal complex of formula Ia and diamagnetic perfluoroalkyl-containing substance, optionally dissolved in an aqueous solvent.

85. **(Previously presented)** A method according to claim 71, wherein the perfluoroalkyl-containing metal complex is in a galenical formulations that contains a paramagnetic, perfluoroalkyl-containing metal complex of formula Ib, and a diamagnetic perfluoroalkyl-containing substance, optionally dissolved in an aqueous solvent.

86. **(Currently amended)** A method according to claim 51, wherein plaque in which contrast agent is uptaken is visualized.

87. **(New)** A method according to claim 51, wherein necroses and tumors in which contrast agent is uptaken are independently and simultaneously visualized.